

S/N Unknown

PATENT

IN THE UNITED STATES PATENT AND TRADEMARK OFFICE

Applicant: Pathiraja A. Gunatillake et al.

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Filed: Herewith

Docket: 1207.008US1

Title: SILOXANE-CONTAINING POLYURETHANE-UREA COMPOSITIONS

PRELIMINARY AMENDMENT

BOX PATENT APPLICATION

Commissioner for Patents

Washington, D.C. 20231

Dear Sir:

Prior to examination of the above-identified patent application, please amend as follows.

IN THE SPECIFICATION

On the first page, after the title, please insert:

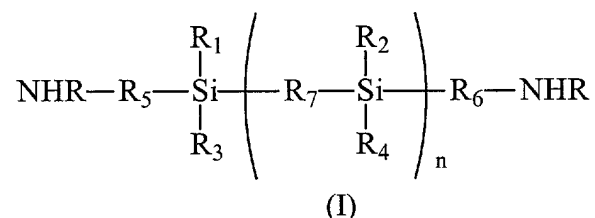
--Cross-Reference to related Applications

This application is a continuation under 35 U.S.C. 111(a) of International Patent Application No. PCT/AU00/00345, filed on April 19, 2000 and published on November 2, 2000 as WO 00/64971, which in turn is an international filing of Australian Application No. PP9917, filed on April 23, 1999, all of which applications are incorporated herein by reference.--

IN THE CLAIMS

Please cancel claims 1-62 and add new claims 63-121 as follows.

63.(NEW) A polyurethane-urea elastomeric composition comprising a soft segment and a hard segment, wherein the soft segment is formed from at least one of a macrodiol, a macrodiamine, and a compound of formula (I):



wherein

R is hydrogen or an optionally substituted straight chain, branched or cyclic, saturated or unsaturated hydrocarbon radical;

R₁, R₂, R₃, R₄, R₅ and R₆ are each independently hydrogen or an optionally substituted straight chain, branched or cyclic, saturated or unsaturated hydrocarbon radical;

R₇ is a divalent linking group or an optionally substituted straight chain, branched or cyclic, saturated or unsaturated hydrocarbon radical; and

n is an integer of 1 or greater;

wherein the macrodiol is a polysiloxane macrodiol, a polyether macrodiol, a polycarbonate macrodiol, or a mixture thereof;

and wherein the hard segment is formed from:

(i) a diisocyanate; and

(ii) a chain extender comprising the compound of formula (I), a second chain extender, or a combination thereof;

provided the soft segment is formed from the compound of formula (I); the hard segment is formed from the compound of formula (I); or the soft segment is formed from the compound of formula (I) and the hard segment is formed from the compound of formula (I).

64.(NEW) The composition of claim 63 wherein n is 1 to 4; the molecular weight of the compound of formula (I) is about 500 or less; and the compound of formula (I) functions as a chain extender.

65.(NEW) The composition of claim 64 wherein R₇ is oxy.

66.(NEW) The composition of claim 64 wherein the compound of formula (I) has a molecular weight range of about 60 to about 500.

67.(NEW) The composition of claim 66 wherein the compound of formula (I) has a molecular weight range of about 60 to about 450.

68.(NEW) The composition of claim 64 wherein the compound of formula (I) is 1,3-bis(3-aminopropyl)tetramethyldisiloxane; or 1,3-bis(4-aminobutyl)tetramethyldisiloxane.

69.(NEW) The composition of claim 64 wherein the chain extender comprises the compound of formula (I) and the second chain extender.

70.(NEW) The composition of claim 69 wherein the second claim extender is a diol, a diamine, a water chain extender, or a combination thereof.

71.(NEW) The composition of claim 70 wherein the diol chain extender is 1,4-butanediol; 1,6-hexanediol; 1,8-octanediol; 1,9-nonanediol; 1,10-decanediol; 1,12-dodecanediol; 1,4-cyclohexanedimethanol; p-xyleneglycol; 1,4 bis (2-hydroxyethoxy) benzene; water; or a combination thereof.

72.(NEW) The composition of claim 70 wherein the diamine chain extender is 1,2-ethylenediamine; 1,3-propanediamine; 1,3-butanediamine; 1,6-hexanediamine; 1,2-diaminocyclohexane; 1,3-diaminocyclohexane; or a combination thereof.

73.(NEW) The composition of claim 69 wherein the compound of formula (I) functions as a chain extender and the molar percentage of the compound of formula (I) is about 1 to about 50% of the composition.

74.(NEW) The composition of claim 69 wherein the compound of formula (I) functions as a chain extender and the molar percentage of the compound of formula (I) is about 35% to about 45% of the composition.

75.(NEW) The composition of claim 63 wherein the diisocyanate is aliphatic or aromatic.

76.(NEW) The composition of claim 75 wherein the diisocyanate is 4,4'-diphenylmethane diisocyanate (MDI); methylene bis (cyclohexyl) diisocyanate (H₁₂MDI); p-phenylene

diisocyanate (p-PDI); trans-cyclohexane-1,4-diisocyanate (CHDI); 1,6-diisocyanatohexane (DICH); 1,5-diisocyanato naphthalene (NDI); para-tetramethylxylene diisocyanate (p-TMXDI); meta-tetramethylxylene diisocyanate (m-TMXDI); 2,4-toluene diisocyanate (2,4-TDI); isophorone diisocyanate (IPDI); an isomer thereof; or a mixture thereof.

77.(NEW) The composition of claim 63 wherein the hard segment is present in about 15 wt.% to about 50 wt.% of the composition.

78.(NEW) The composition of claim 77 wherein the hard segment is present in about 21.8% to about 50 wt.% of the composition.

79.(NEW) The composition of claim 77 wherein the hard segment is present in about 21.8% to about 40 wt.% of the composition.

80.(NEW) The composition of claim 63 wherein the soft segment is formed from a polysiloxane macrodiol, a polyether macrodiol, a polyether macrodiamine, or a mixture thereof.

81.(NEW) The composition of claim 63 wherein n is about 5 to about 100; the molecular weight of the compound of formula (I) is about 500 to about 10,000; and the soft segment is formed from the compound of formula (I).

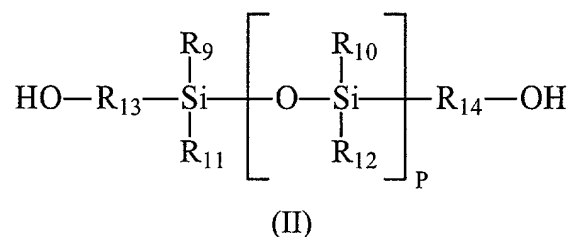
82.(NEW) The composition of claim 81 wherein the compound of formula (I) is an amine-terminated PDMS.

83.(NEW) The composition of claim 82 wherein the amine-terminated PDMS is bis(3-aminopropyl)-polydimethyl siloxane.

84.(NEW) The composition of claim 81 wherein the soft segment is formed from the compound of formula (I) and at least one of a macrodiol and a macrodiamine.

85.(NEW) The composition of claim 84 wherein the macrodiol is a polysiloxane macrodiol, a polyether macrodiol, a polyester macrodiol, a polycarbonate macrodiol, or a mixture thereof.

86.(NEW) The composition of claim 85 wherein the polysiloxane macrodiol is a compound of formula (II):



wherein

R_9 , R_{10} , R_{11} , R_{12} , R_{13} and R_{14} are each independently an optionally substituted straight chain, branched or cyclic, saturated or unsaturated hydrocarbon radical; and

p is an integer of 1 to about 100.

87.(NEW) The composition of claim 86 wherein the compound of formula (II) is PDMS.

88.(NEW) The composition of claim 87 wherein R_{13} and R_{14} are each independently propylene, butylene, pentylene, hexylene, ethoxypropyl, propoxypropyl, or butoxypropyl.

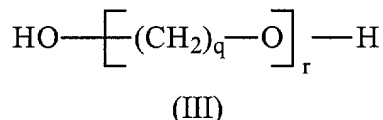
89.(NEW) The composition of claim 86 wherein the molecular weight of the compound of formula (II) is about 200 to about 6,000.

90.(NEW) The composition of claim 86 wherein the molecular weight of the compound of formula (II) is about 500 to about 2,000.

91.(NEW) The composition of claim 84 wherein the soft segment is formed from an amine-terminated PDMS and PDMS.

92.(NEW) The composition of claim 85 wherein the polyether macrodiol is a compound of

formula (III).



wherein

q is an integer of 4 or more; and

r is an integer of 2 to 50.

93.(NEW) The composition of claim 92 wherein q is about 5 or higher.

94.(NEW) The composition of claim 93 wherein the compound of formula (III) is poly(hexamethylene oxide) (PHMO); poly(heptamethylene oxide); poly(octamethylene oxide) (POMO); or poly(decamethylene oxide) (PDMO).

95.(NEW) The composition of claim 92 wherein the soft segment is formed from the compound formula (I), which functions as the macrodiamine; and the compound of formula (III), which functions as a macrodiol.

96.(NEW) The composition of claim 85 wherein the molecular weight of the compound of formula (III) is about 200 to about 5,000.

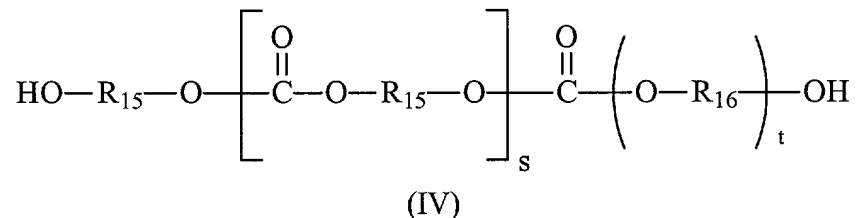
97.(NEW) The composition of claim 96 wherein the molecular weight of the compound of formula (III) is about 500 to about 1,200.

98.(NEW) The composition of claim 85 wherein the polycarbonate macrodiol is a poly(alkylene carbonate); a polycarbonate prepared by reacting an alkylene carbonate with an alkanediol; a silicon based polycarbonate prepared by reacting an alkylene carbonate with 1,3-bis(4-hydroxybutyl)-1,1,3,3-tetramethyldisiloxane (BHTD); an alkanediol; or a mixture thereof.

99.(NEW) The composition of claim 85 wherein both the polyether macrodiol and the

polycarbonate macrodiol are present as a mixture or a copolymer.

100.(NEW) The composition of claim 99 wherein the copolymer is a copoly(ether carbonate) macrodiol represented by the compound of formula (IV):



wherein

R_{15} and R_{16} are each independently an optionally substituted straight chain, branched or cyclic, saturated or unsaturated hydrocarbon radical; and s and t are integers of 1 to about 20.

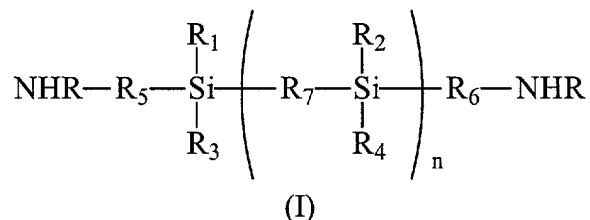
101.(NEW) The composition of claim 84 wherein the macrodiamine is a polyether macrodiamine.

102.(NEW) The composition of claim 101 wherein the polyether macrodiamine is POLAMINE 650.

103.(NEW) The composition of claim 63 wherein the soft segment is formed from the compound of formula (I) and at least one of a macrodiol and a macrodiamine.

104.(NEW) A polyurethane-urea elastomeric composition comprising a soft segment and a hard segment, wherein the soft segment is formed from:

- (i) a macrodiol comprising a polysiloxane macrodiol and a polyether macrodiol; and wherein the hard segment is formed from:
- (ii) a diisocyanate; and
- (iii) a chain extender selected from the group consisting of a compound of formula (I):



wherein

R is hydrogen or an optionally substituted straight chain, branched or cyclic, saturated or unsaturated hydrocarbon radical;

R₁, R₂, R₃, R₄, R₅ and R₆ are each independently hydrogen or an optionally substituted straight chain, branched or cyclic, saturated or unsaturated hydrocarbon radical;

R₇ is a divalent linking group or an optionally substituted straight chain, branched or cyclic, saturated or unsaturated hydrocarbon radical; and

n is an integer of 1 to about 4;

the molecular weight of the compound of formula (I) is about 500 or less;

a diamine chain extender; 1,3-bis(3-aminopropyl)tetramethyldisiloxane; 1,3-bis(4-aminobutyl)tetramethyldisiloxane; 1,4-butanediol; 1,2-ethylenediamine; ethanolamine; hexamethylenediamine; 1,4-butanediamine; water; 1,4-bis(4-hydroxybutyl)tetramethyldisiloxane; and combinations thereof.

105.(NEW) The composition of claim 104 wherein the weight ratio of polysiloxane macrodiol to polyether macrodiol in the composition is about 1:99 to about 99:1.

106.(NEW) The composition of claim 104 wherein the weight ratio of polysiloxane to polyether is about 75:25 to about 85:15.

107.(NEW) The composition of claim 102 wherein the weight percentage of the macrodiol in the composition is about 60 wt.% to about 40 wt.%.

108.(NEW) The composition of claim 104 wherein the diisocyanate is MDI.

109.(NEW) A polyurethane-urea elastomeric composition comprising a soft segment and a hard segment, wherein the soft segment is formed from:

(i) a polysiloxane macrodiamine; and either a polyether macrodiol or a polyether macrodiamine;

and wherein the hard segment is formed from:

(ii) a diisocyanate; and

(iii) a diamine chain extender, 1,3-bis(3-aminopropyl)tetramethyldisiloxane; 1,3-bis(4-aminobutyl)tetramethyldisiloxane; 1,4-butanediol; 1,2-ethylenediamine; ethanolamine; hexamethylenediamine; 1,3-diaminocyclohexane; 1,2-diamino cyclohexane; water; 1,3-bis(4-hydroxybutyl) 1,1,3,3-tetramethyldisiloxane; or a combination thereof.

110.(NEW) The composition of claim 109 wherein the amount of hard segment in the composition is about 15 wt.% to about 50 wt.%.

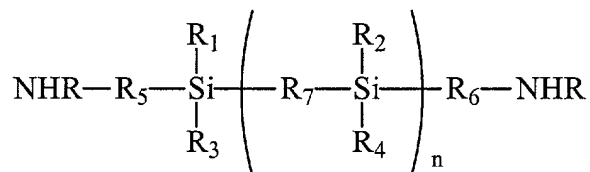
111.(NEW) A polyurethane-urea elastomeric composition comprising a soft segment and a hard segment, wherein the soft segment is formed from:

(i) a macrodiol or a macrodiamine selected from the group consisting of a polysiloxane macrodiol, a polyether macrodiol, a polyester macrodiol, a polycarbonate macrodiol; a polyether macrodiamine; and mixtures thereof;

and wherein the hard segment is formed from:

(ii) a diisocyanate; and

(iii) a chain extender comprising a compound of formula (I):



wherein

R is hydrogen or an optionally substituted straight chain, branched or cyclic, saturated or unsaturated hydrocarbon radical;

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R_1 , R_2 , R_3 , R_4 , R_5 and R_6 are each independently hydrogen or an optionally substituted straight chain, branched or cyclic, saturated or unsaturated hydrocarbon radical;

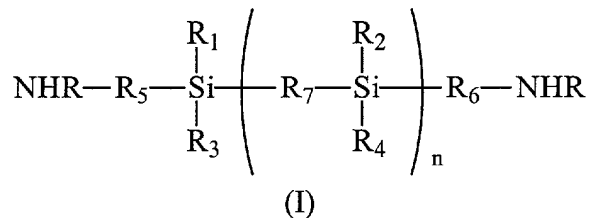
R_7 is a divalent linking group or an optionally substituted straight chain, branched or cyclic, saturated or unsaturated hydrocarbon radical; and

n is an integer of 1 to about 4; and

the compound of formula (I) has a molecular weight of about 500 or less.

112.(NEW) A polyurethane-urea elastomeric composition comprising a soft segment and a hard segment, wherein the soft segment is formed from:

- (i) a macrodiol comprising a polysiloxane macrodiol and a polycarbonate macrodiol; and the hard segment is formed from:
- (ii) a diisocyanate; and
- (iii) chain extender selected from the group consisting of a compound of formula (I):



wherein

R is hydrogen or an optionally substituted straight chain, branched or cyclic, saturated or unsaturated hydrocarbon radical;

R_1 , R_2 , R_3 , R_4 , R_5 and R_6 are each independently hydrogen or an optionally substituted straight chain, branched or cyclic, saturated or unsaturated hydrocarbon radical;

R_7 is a divalent linking group or an optionally substituted straight chain, branched or cyclic, saturated or unsaturated hydrocarbon radical; and

n is an integer of 1 to about 4; and

the compound of formula (I) has a molecular weight of about 500 or less;

a diamine chain extender; 1,3-bis(3-aminopropyl)tetramethyldisiloxane; 1,3-bis(4-aminobutyl)tetramethyldisiloxane; 1,4-butanediol; 1,2-ethylenediamine; ethanolamine;

hexamethylenediamine; 1,4-butanediamine; water 1,4-bis(4-hydroxybutyl)tetramethyldisiloxane; and combinations thereof;

wherein the level of hard segment in the composition is about 21.8 wt.% to about 60 wt.%.

113.(NEW) The composition of claim 63 having improved mechanical properties, clarity, processability or degradation resistance.

114.(NEW) The composition of claim 63 that is resistant to cyclic flex fatigue.

115.(NEW) The composition of claim 63 that is resistant to degradation.

116.(NEW) The composition of claim 63 that is resistant to degradation *in vivo*.

117.(NEW) The composition of claim 63 that is useful as a biomaterial.

118.(NEW) A medical device, articles or implants composed wholly or partly of the composition of claim 63.

119.(NEW) The medical device, articles or implant of claim 118 which is a cardiac pacemaker, defibrillator, electromedical device, catheters, cannula, implantable prostheses, cardiac assist device, heart valve, vein valve, vascular graft, extra-corporeal device, artificial organ, pacemaker lead, defibrillator lead, blood pump, balloon pump, A-V shunt, biosensor, membranes for cell encapsulation, drug delivery device, wound dressing, artificial joint, orthopaedic implant, or soft tissue replacement.

120.(NEW) A device or article composed wholly or partly of the composition of claim 63.

121.(NEW) The device or article of claim 120 which is artificial leather, a shoe sole; cable sheathing; varnish, coating; structural components for a pump, vehicle; mining ore screen,

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conveyor belt; laminating compound, textile; separation membrane; sealants or a component of an adhesive.

REMARKS

The claims have been amended to comply with standard U.S. practice. For example, the multiple dependencies have been removed. As such, the amendments do not limit the full scope of the invention, as originally claimed.

Respectfully submitted,

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By their Representatives,

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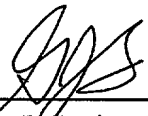
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